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$^{243}\text{Am} + ^{48}\text{Ca}$: A Second Look at the Rf/Db Data Set
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In December of 2005 a series of experiments were performed at the U400 Cyclotron at the Flerov Laboratory of Nuclear Reactions in Dubna, Russia, using the reaction $^{243}\text{Am} (^{48}\text{Ca}, 3n) ^{288}115$ which, after a sequence of five alpha decays, produces a long-lived (~1d) fission activity that had been detected previously in experiments using the Dubna Gas Filled Recoil Separator (DGFRS). These experiments were attempts to establish the elemental identity of the fissioning species as dubnium (element 105), which in turn confirms the identity of the original parent nucleus as element 115 through genetic correlation of the subsequent alpha decays. A series of approximately 24-hour bombardments were followed by chemical separations designed to isolate the Group Four and Five chemical fractions, and then separate the Nb and Ta fractions, which are Group Five homologues of dubnium and should therefore behave chemically similar. The samples were prepared for alpha and fission measurement and counted for an extended period of time [1,2]. Fission events were detected in the Ta-like fractions only, which correspond to the fission coming from either the ^{268}Db isotope directly or long-lived electron-capture decay in ^{268}Db followed by a short half-life fission of ^{268}Rf .

In May of 2007 the Rf and Db fractions were recounted for very long times on alpha spectrometers to look at what species remained after approximately 1.5 years of time. One of the issues to be resolved was the potential for actinide contamination of the counting samples, which might have adverse affects on the observed data from the original experiment. In the original experiment the samples had significant quantities of β - γ activity which made the absolute identification of the alpha activity in each sample difficult. By allowing the β - γ activity to decay away, it gives us the opportunity to make definitive identifications of any alpha emitting isotopes on the sample planchets.

The current set of alpha spectra revealed the presence of; ^{243}Am , which is knock-off activity from the target; ^{241}Am , which is a contaminant in the target material; and $^{242}\text{Cm} + ^{243,244}\text{Cm}$, which are transfer products. ^{208}Po was also seen in the spectra, which would be produced by transfer reactions on Pb impurities in the target or target backing.

In general the Nb fractions showed consistent contamination with ^{243}Am , ranging from 3.3 to 5.0 dpm in the measured sample. Also noted in the Nb fractions was a significant amount of ^{208}Po , ranging from 2.9 to 5.2 dpm in the measured sample. The alpha data is shown in Table 1.

Using the measured alpha-activity of the observed isotopes ^{241}Am , ^{243}Am , ^{242}Cm , and $^{243+244}\text{Cm}$ and applying the fission branching ratio for each isotope we are able to calculate the fissions per day rate, corrected to the time of the experiment. Since ^{243}Cm and ^{244}Cm would be largely indistinguishable in the samples, we also applied the ^{244}Cm

fission branch to the ^{243}Cm alpha activity to help establish an upper limit based upon the observed alpha activity. This data is displayed in Table 2.

Within the recount data, generally the highest actinide contamination was found in the Nb fractions, with the noted exception of the Ta fractions from 12/12/05 and 12/18/05. The 12/18/05 Ta fraction did not have a detected fission event in it. Looking at the combined fission rate of the detected nuclides in the 12/12/05 Ta fraction, which is $1.01\text{E-}3$ per day, it is quite unlikely that the observed fission came from the measured actinide activity.

α - dpm - background corrected												
PSI-given Sample ID	²⁰⁸ Po	MDA	²⁴² Cm	MDA	²⁴³ Cm	MDA	²⁴¹ Am	MDA	²⁴³ Am	MDA	Det	Count Time (s)
12/10/05 Nb - JINR	5.24E+00	3.95E-03	1.40E-02	3.97E-03	1.46E-02	3.95E-03	2.51E-04	3.95E-03	5.05E+00	3.95E-03	2a	132950.2
12/10/05 Ta - LLNL	1.21E-02	1.03E-02	1.31E-02	1.03E-02	1.06E-02	1.80E-03	1.23E-02	1.87E-02	-3.09E-02	1.38E-02	2b	292000.0
12/12/05 Ta - JINR	3.18E-01	3.60E-02	1.19E+00	1.57E-02	4.84E-01	3.49E-03	1.98E-01	1.56E-02	9.35E-01	3.69E-02	2b	150570.9
12/14/05 Nb - JINR	2.94E+00	3.95E-03	6.04E-02	3.97E-03	2.48E-02	3.95E-03	7.53E-03	3.95E-03	3.32E+00	3.95E-03	2b	132946.3
12/14/05 Ta - LLNL	-5.84E-03	2.98E-02	4.44E-03	1.23E-02	9.31E-03	1.80E-03	8.35E-03	1.22E-02	-6.75E-03	3.06E-02	2b	292000.0
12/18/05 Nb - JINR	3.03E+00	2.70E-03	1.55E-02	2.72E-03	5.29E-02	2.70E-03	2.86E-02	2.70E-03	3.66E+00	2.70E-03	2a	194472.8
12/18/05 Ta - JINR	2.98E+00	1.90E-02	1.15E-01	1.90E-02	2.78E-01	6.95E-03	1.17E-01	3.11E-02	5.31E+00	2.40E-02	2a	75583.5
12/20/05 Nb - JINR	3.78E+00	2.70E-03	8.67E-02	2.72E-03	3.10E-02	2.70E-03	5.37E-03	2.70E-03	4.47E+00	2.70E-03	2b	194517.3
12/20/05 Ta - JINR	4.68E-01	4.72E-02	1.10E-01	2.20E-02	4.55E-02	6.95E-03	1.62E-02	2.19E-02	5.76E-01	4.83E-02	2b	205105.6

Table 1 – Alpha count data for the measured samples. The Sample ID laboratory designation is arbitrary. The chemical yield of the procedure is not taken into account in the results shown. MDA represents the minimum detectable amount of the specific nuclide.

fissions per day at experiment time							
PSI-given Sample ID	²⁴² Cm	²⁴³ Cm	²⁴¹ Am	²⁴³ Am	If ²⁴³ Cm was ²⁴⁴ Cm	Total Fission Rate	Fission?
12/10/05 Nb - JINR	1.20E-05	1.16E-09	1.56E-12	2.69E-07	3.05E-05	4.27E-05	No
12/10/05 Ta - LLNL	1.15E-05	8.43E-10	7.66E-11	-1.65E-09	2.22E-05	3.37E-05	Yes (30h)
12/12/05 Ta - JINR	9.94E-04	3.83E-08	1.23E-09	4.98E-08	1.01E-03	2.00E-03	Yes (27h)
12/14/05 Nb - JINR	5.08E-05	1.97E-09	4.68E-11	1.77E-07	5.18E-05	1.03E-04	No
12/14/05 Ta - LLNL	3.81E-06	7.38E-10	5.19E-11	-3.60E-10	1.94E-05	2.32E-05	Yes (16 h)
12/18/05 Nb - JINR	1.27E-05	4.19E-09	1.77E-10	1.95E-07	1.10E-04	1.23E-04	No
12/18/05 Ta - JINR	9.21E-05	2.20E-08	7.24E-10	2.83E-07	5.78E-04	6.70E-04	No
12/20/05 Nb - JINR	7.05E-05	2.45E-09	3.33E-11	2.38E-07	6.45E-05	1.35E-04	No
12/20/05 Ta - JINR	8.75E-05	3.60E-09	1.00E-10	3.07E-08	9.47E-05	1.82E-04	Yes (18 h)

Table 2 – Fission data calculated using the fission branching ratios for the listed nuclides. The Total Fission Rate is in the units of fissions per day, and is a sum of the results shown. The rates are decay-corrected to the time of the experiment shown in the Sample ID field.

Using the measured activity as a scalar we can calculate other shorter half-life actinide nuclides that would be produced by transfer reaction and determine their contribution to the fission rate in the samples. In doing this, we assumed that the transfer of mass from the projectile to the target was similar in magnitude as that observed in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction, and that the distribution of charge in the final products was consistent with the number of emitted neutrons calculated based on relative reaction Q-values. While we expect no contribution to the fission rate from the odd-proton nuclides, we calculate that the SF decay rate due to heavy californium isotopes during the first day after the end of the irradiation + chemical separation was about twice that due to the curium isotopes (assuming similar chemical yields), about half of it from decays of ^{252}Cf . The major contributor to the SF rate of the sample in the fermium isotopes was ^{256}Fm (2.63 h), which had an instantaneous decay rate of 0.032 per day immediately after the completion of chemistry if scaled to the worst-case Ta sample. No other radionuclides contributed to the SF decay rate of the sample. Based on these data, spontaneous fission decays of the actinide transfer products clearly did not contribute significantly to the observed fissions in the chemical fractions, which means that the observed fission activities were from the decay of element 105.

References:

- [1] "Separation of Group Five Elements by Reversed-Phase Chromatography", P.A. Wilk, J.H. Landrum, D.A. Shaughnessy, J.M. Kenneally, N.J. Stoyer, M.A. Stoyer, K.J. Moody, N.V. Aksenov, G.A. Bozhikov, E.E. Tereshatov, G.K. Vostokin, S.V. Shishkin, and S.N. Dmitriev, J. Radioanal. Nucl. Chem. 275, 651657, 2008, UCRL-JRNL-228727.
- [2] "Chemical Identification of a Long-Lived Isotope of Dubnium, a Descendant of Element 115", N.J. Stoyer, J.H. Landrum, P.A. Wilk, K.J. Moody, J.M. Kenneally, D.A. Shaughnessy, M.A. Stoyer, J.F. Wild, R.W. Loughheed, S.N. Dmitriev, Yu.Ts. Oganessian, S.V. Shishkin, N.V. Aksenov, E.E. Tereshatov, G.A. Bozhikov, G.K. Vostokin, V.K. Utyonkov, and A.V. Yerebin, Nucl. Phys. A 787, 388C, 2007, UCRL-PROC-224777.